

FORM PTO-1390) U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE (REV. 9-2001)		ATTORNEY'S DOCKET NUMBER 355.40958X00 filed December 20, 2001
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		U.S. APPLICATION NO. (if known, see 37 CFR 1.5) <div style="font-size: 24pt; font-weight: bold;">10/018531</div>
INTERNATIONAL APPLICATION NO. PCT/GB00/02536	INTERNATIONAL FILING DATE June 30, 2000	PRIORITY DATE CLAIMED July 2, 1999
TITLE OF INVENTION ELECTROLYTE REBALANCING SYSTEM		
APPLICANT(S) FOR DO/EO/US MORRISSEY, PATRICK JOHN MITCHELL, PHILIP JOHN MALE, STEWART ERNEST		
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:		
<ol style="list-style-type: none"> 1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. 3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below. 4. <input checked="" type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31). 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)) <ol style="list-style-type: none"> a. <input type="checkbox"/> is transmitted hereto (required only if not communicated by the International Bureau). b. <input checked="" type="checkbox"/> has been communicated by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office(RO/US) 6. <input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). <ol style="list-style-type: none"> a. <input checked="" type="checkbox"/> is attached hereto. b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4). 7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) <ol style="list-style-type: none"> a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> have been communicated by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). 10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). <p>Items 11 to 20 below concern document(s) or information included:</p> <ol style="list-style-type: none"> 11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 13. <input checked="" type="checkbox"/> A FIRST preliminary amendment. 14. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. 15. <input type="checkbox"/> A substitute specification. 16. <input checked="" type="checkbox"/> A change of power of attorney and/or address letter. 17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825. 18. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4). 19. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4). 20. <input checked="" type="checkbox"/> Other items or information: PCT Request Form; International Publication No. WO 01/03221; International Preliminary Examination Report; International Search Report, Credit Card Payment Form,; Figs 1A-B, 2-8 		

U.S. APPLICATION NO. (If known, see 37 CFR 1.53) 107018551		INTERNATIONAL APPLICATION NO. PCT/GB00/02536		ATTORNEY'S DOCKET NUMBER 355.40958X00	
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21. The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492(a) (1) - (5)):

☐ Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO.....\$1040.00

☒ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO.....\$890.00

☐ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO.....\$740.00

☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4).....\$710.00

☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4).....\$100.00

ENTER APPROPRIATE BASIC FEE AMOUNT =

Surcharge of \$130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492(e)).

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$
Total Claims	- 20 =		x \$18.00	\$0.00
Independent Claims	- 3 =		x \$84.00	\$84.00
MULTIPLE DEPENDENT CLAIMS(S) (if applicable)			+ \$280.00	\$0.00
TOTAL OF ABOVE CALCULATIONS =				\$974.00
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				\$0.00
SUBTOTAL =				\$974.00
Processing fee of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$0.00
TOTAL NATIONAL FEE =				\$974.00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				\$0.00
TOTAL FEES ENCLOSED =				\$974.00

Amount to be refunded:

charged:

CALCULATIONS PTO USE ONLY

a. ☐ A check in the amount of \$_____ to cover the fees is enclosed.

b. ☐ Please charge my Deposit Account No. **01-2135** in the amount of \$_____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. **01-2135**. A duplicate copy of this sheet is enclosed.

d. ☒ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

Antonelli, Terry, Stout & Kraus, LLP
 1300 North Seventeenth Street
 Suite 1800
 Arlington, VA 22209
 USA

SIGNATURE
 Alan E. Schiavelli
 NAME
 32,087
 REGISTRATION NO.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: MORRISSEY et al
Serial No.:
Filed: December 20, 2001
For: Electrolyte Rebalancing System
Group:
Examiner:

PRELIMINARY AMENDMENT

Assistant Commissioner
for Patents
Washington, D.C. 20231

December 20, 2001

Sir:

Prior to examination on the merits of this application and prior to calculation of the filing fee, please amend the above-identified application as follows:

IN THE CLAIMS:

Please amend the claims to read as follows:

6. (Amended) A process as claimed in claim 3 wherein the electrolyte circulating through the -^{ve} chamber of the auxiliary cell during rebalancing is a fraction of electrolyte 2 and wherein the fraction is subsequently treated to remove sulfate ions contained therein.
8. (Amended) A process as claimed in claim 2 wherein the fraction of electrolyte 1 or 2 which is circulated through the -^{ve} chamber of the auxiliary cell is returned to the main stream of electrolyte 1 or 2 respectively.
9. (Amended) A process as claimed in claim 1 which additionally comprises adding

elemental sulfur and/or a sulfide salt to electrolyte 1 in an amount sufficient to restore the initial concentration of sulfur species.

14. (Amended) Apparatus as claimed in claim 12 which additionally comprises a second auxiliary cell which comprises a $+^{ve}$ chamber containing an inert $+^{ve}$ electrode and a $-^{ve}$ chamber containing an inert $-^{ve}$ electrode, the chambers being separated from one another by a cation exchange membrane, means for circulating a fraction of electrolyte 1 or 2 through the $+^{ve}$ chamber and means for circulating a fraction of electrolyte 1 or 2 through the $-^{ve}$ chamber.

16. (Amended) Apparatus as claimed in claim 12 wherein the electrolyte circulated through the $-^{ve}$ chamber of the auxiliary cell is electrolyte 2, additionally comprising means for removing sulfate ions from the fraction of electrolyte 2 after circulation through the $-^{ve}$ chamber of the auxiliary cell.

18. (Amended) Apparatus as claimed in claim 11 additionally comprising means for passing the fraction of electrolyte 1 or 2 which is circulated through the $-^{ve}$ chamber of the auxiliary cell back to the main stream of electrolyte 1 or 2 respectively.

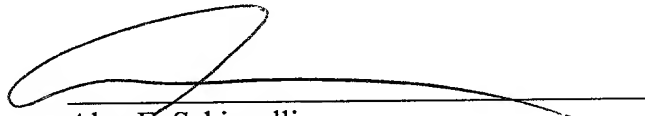
REMARKS

The foregoing amendments are respectfully requested prior to examination on the merits of this application. A marked up copy of the amended claims is attached.

To the extent necessary, applicants petition for an extension of time under 37 CFR 1.136. Please charge any shortage in the fees due in connection with the filing of this paper, including extension of time fees, to the deposit account of Antonelli, Terry, Stout & Kraus, LLP, Deposit Account No. 01-2135 (Case: 355.40958X00), and please credit any excess fees to such deposit account.

Respectfully submitted,

ANTONELLI, TERRY, STOUT & KRAUS, LLP



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Registration No. 32,087

AES/jla
(703) 312-6600

REWRITTEN MARKED UP COPY

6. (Amended) A process as claimed in ~~any one of claims 3 to 5~~ claim 3 wherein the electrolyte circulating through the ^{-ve} chamber of the auxiliary cell during rebalancing is a fraction of electrolyte 2 and wherein the fraction is subsequently treated to remove sulfate ions contained therein.

8. (Amended) A process as claimed in ~~any one of claims 2 to 7~~ claim 2 wherein the fraction of electrolyte 1 or 2 which is circulated through the ^{-ve} chamber of the auxiliary cell is returned to the main stream of electrolyte 1 or 2 respectively.

9. (Amended) A process as claimed in ~~any one of the preceding claims~~ claim 1 which additionally comprises adding elemental sulfur and/or a sulfide salt to electrolyte 1 in an amount sufficient to restore the initial concentration of sulfur species.

14. (Amended) Apparatus as claimed in claim 12 ~~or claim 13~~ which additionally comprises a second auxiliary cell which comprises a ^{+ve} chamber containing an inert ^{+ve} electrode and a ^{-ve} chamber containing an inert ^{-ve} electrode, the chambers being separated from one another by a cation exchange membrane, means for circulating a fraction of electrolyte 1 or 2 through the ^{+ve} chamber and means for circulating a fraction of electrolyte 1 or 2 through the ^{-ve} chamber.

16. (Amended) Apparatus as claimed in ~~any one of claims 12 to 15~~ claim 12 wherein the electrolyte circulated through the ^{-ve} chamber of the auxiliary cell is electrolyte 2, additionally comprising means for removing sulfate ions from the fraction of electrolyte 2 after circulation through the ^{-ve} chamber of the auxiliary cell.

18. (Amended) Apparatus as claimed in ~~any one of claims 11 to 17~~ claim 11 additionally comprising means for passing the fraction of electrolyte 1 or 2 which is circulated through the ^{-ve} chamber of the auxiliary cell back to the main stream of

electrolyte 1 or 2 respectively.

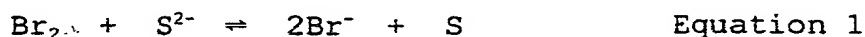
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Electrolyte Rebalancing System

The present invention relates to the field of regenerative fuel cell (RFC) technology. In particular it relates to apparatus and methods for the operation of RFCs which enhance their performance characteristics.

The manner in which RFCs are able to store and deliver electricity is well known to those skilled in the art. An example of an RFC is described in US-A-4485154 which discloses an electrically chargeable, anionically active, reduction-oxidation system using a sulfide/polysulfide reaction in one half of the cell and an iodine/iodide, chlorine/chloride or bromine/bromide reaction in the other half of the cell. The two halves of the cell are separated by a cation exchange membrane.

The overall chemical reaction involved, for example, for the bromine/bromide-sulfide/polysulfide system is shown in Equation 1 below:



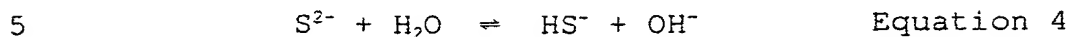
However, within an RFC such as that described in US-A-4485154, the reaction takes place in separate but dependent bromine and sulfur half-cell reactions as shown below in Equations 2 and 3:



It should be noted however that these equations represent the overall reactive changes occurring in the RFC. In practice the reactions are complicated by

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the low basicity of sulfide which results in the formation of bisulfide as the active species, as shown in Equation 4.



Also, the sulfur produced in Equations 1 and 3 forms soluble polysulfide species in the presence of sulfide ions, as shown in Equation 5 (where x may be from 1 to 4).



Also, free bromine is solubilised in the presence of bromide ions to form the tribromide ion, as shown in Equation 6



20 When the RFC is discharging, bromine is converted to bromide on the +^{ve} side of the membrane and sulfide is converted to polysulfide on the -^{ve} side of the membrane. Equation 1 goes from left to right and metal ions flow from the -^{ve} side of the membrane to the +^{ve} side of the membrane to complete the circuit. When the RFC is charging, bromide is converted to bromine on the +^{ve} side of the membrane and polysulfide is converted to sulfide on the -^{ve} side of the membrane. Equation 1 goes from right to left and metal ions flow from the +^{ve} side of the membrane to the -^{ve} side of the membrane to complete the circuit.

30 The discharge/charge cycle described above will be repeated many times during the lifetime of the RFC and in order for the RFC to work efficiently throughout its lifetime it is important that the electrolytes remain balanced. In the context of the present

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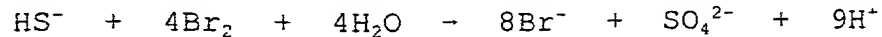
specification, when the term "balanced" is used to describe the electrolytes it means that the relative concentrations of the reactive species within the electrolytes are maintained at, or close to, values which enable optimum performance of the RFC. Similarly, in the context of the present specification, the term "rebalancing" refers to a process which alters the concentration of one or more reactive species in one or both of the electrolytes so as to return said electrolytes to a balanced state or so as to maintain said electrolytes in a balanced state.

At the beginning of the RFC's lifetime the relative concentrations of the reactive species on either side of the membrane will normally be fixed so that the electrolytes are balanced. However, once the RFC begins to operate in its repeating discharge-charge cycle, factors may intervene which result in the electrolytes becoming unbalanced. These factors will vary depending upon the identity of the reactive species within the electrolytes and on the manner in which the RFC is constructed and operated.

In the case of the bromine/bromide-sulfide/polysulfide RFC such as that described above, the most important factor which results in the electrolytes becoming unbalanced is the diffusion of unwanted species across the membrane. Although a cation selective ion-exchange membrane is used, 100% permselectivity is not possible and during extended cycling of the cell some anionic species diffuse through the membrane. In particular, sulfide ions (largely present in the bisulfide form, HS^-) and polysulfide ions (S_{x+1}^{2-} , where x may be from 1 to 4) may diffuse from the sulfide/polysulfide electrolyte into the bromine/bromide electrolyte where they will be

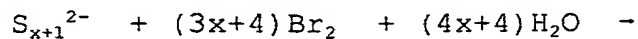
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oxidised by the bromine to form sulfate ions as shown in equations 7 and 8 below:

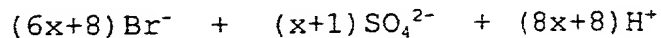


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Equation 7



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Equation 8

Imperfections other than diffusion through the membrane which could similarly contribute to the above process are ineffective sealing between cell compartments, or catastrophic failure of any of the cell separating components, each of which may result in crossover of the electrolytes between cell compartments.

In Equations 7 and 8, the oxidation of the sulfur species goes beyond that which occurs during normal operation of the RFC. That is to say, the sulfide and polysulfide ions are oxidised all the way to sulfate ions. Consequently, in the case of sulfide ion crossover (Equation 7), four bromine molecules per sulfide ion are consumed rather than the normal one bromine molecule per sulfide ion which is consumed in the reaction scheme of Equation 1. Similar overconsumption of bromine results from polysulfide cross-over (Equation 8) although to a slightly lesser extent. As a result, the bromine/bromide electrolyte becomes discharged to a greater extent than the sulfide/polysulfide electrolyte. Thus, when the cell is discharging there is insufficient bromine present to react with all the sulfide ions present thereby

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preventing completion of the discharge cycle. As a result, the voltage generated by the cell begins to decline earlier in the discharge cycle than when the electrolytes are balanced. In effect, the reactions represented by Equations 7 and 8 result in the conversion of some of the polysulfide ions to sulfide because not all of the polysulfide ions are recovered on discharge. Subsequent cycles repeat this process, further reducing the number of polysulfide ions present. Ultimately, there will be insufficient polysulfide ions present to accept charge during the charge cycle. Since the electrochemistry has to continue if charging is maintained, the next most favourable reaction occurs, i.e. water is reduced and the electrode on the $-ve$ side of the cell starts to gas hydrogen.

It would therefore be advantageous to provide a process for rebalancing the electrolytes in order to compensate for the unbalancing effect of the crossover of sulfide and/or polysulfide electrolyte species into the bromine electrolyte. Although it would be possible to replace the electrolytes in the system with fresh electrolytes at periodic intervals, this is disadvantageous because of the economic implications and because of the environmental implications of the great amounts of waste electrolytes which would require to be disposed of.

Accordingly, the present invention provides an electrochemical process for energy storage and/or power delivery comprising:

- (i) maintaining and circulating electrolyte flows in a fully liquid system in which the active constituents are fully soluble in a single cell or in an array of repeating cell structures, each cell with a positive ($+ve$) chamber containing an

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- inert +^{ve} electrode and a negative (-^{ve}) chamber containing an inert -^{ve} electrode, the chambers being separated from one another by a cation exchange membrane, the electrolyte circulating in the -^{ve} chamber of each cell during power delivery containing a sulfide (electrolyte 1), and the electrolyte circulating in the +^{ve} chamber during power delivery containing bromine (electrolyte 2),
- (ii) restoring or replenishing the electrolytes in the +^{ve} and -^{ve} chambers by circulating the electrolyte from each chamber to storage means comprising a volume of electrolyte greater than the cell volume for extended delivery of power over a longer discharge cycle than the cell volume alone would permit, and
- (iii) rebalancing the electrolytes by circulating a fraction of electrolyte 1 or electrolyte 2 through the +^{ve} chamber of an auxiliary cell, said auxiliary cell comprising a +^{ve} chamber containing an inert +^{ve} electrode and a -^{ve} chamber containing an inert -^{ve} electrode, the chambers being separated from one another by a cation exchange membrane, the electrolyte circulating through the -^{ve} chamber of the auxiliary cell containing water and being free from polysulfide and free from bromine during rebalancing, the auxiliary cell operating so as to oxidise sulfide ions to sulfur or bromide ions to bromine in the +^{ve} chamber and so as to reduce water to hydrogen and hydroxide ions in the -^{ve} chamber.

The oxidation of bromide to bromine rebalances the electrolytes by restoring the bromine which is reduced by reaction with migrating sulfide ions. Oxidation of bromide to bromine may also be thought of as charging

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the bromine/bromide electrolyte since the chemical content of the bromine/bromide electrolyte changes in the same manner as when the RFC is in its charging cycle.

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The oxidation of sulfide to sulfur rebalances the electrolytes by oxidising the equivalent amount of sulfide which would ordinarily have been oxidised by the halogen which was reduced by reaction with migrating sulfide ions. Oxidation of polysulfide to sulfur may also be thought of as discharging the sulfide/polysulfide electrolyte since the chemical content of the sulfide/polysulfide electrolyte changes in the same manner as in the RFC when it is in its discharging cycle.

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In order that rebalancing of the electrolytes may occur, it is essential that during the rebalancing process the electrolyte circulating through the $-ve$ chamber of the auxiliary cell should be free from polysulfide and free from bromine. The reason for this is that these chemical species are more readily reduced than water. If electrolyte 1 is circulated through the $+ve$ chamber of the auxiliary cell and the electrolyte circulating through the $-ve$ chamber of the auxiliary cell contains polysulfide, then the reaction which will occur in the $-ve$ chamber will be reduction of polysulfide to sulfide rather than reduction of water to hydrogen and hydroxide ions. This would result in no net change in the oxidation state of the sulfur species present in the system. If reduction of water is to occur in the presence of polysulfide the $-ve$ electrode in the $-ve$ chamber must be specially constructed to starve it of polysulfide. Similarly, if electrolyte 2 is circulated through the $+ve$ chamber of the auxiliary cell and the electrolyte circulating through the $-ve$ chamber of the auxiliary cell contains

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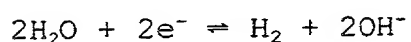
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bromine, then the reaction which will occur in the ^{-ve} chamber will be reduction of bromine to bromide rather than reduction of water to hydrogen and hydroxide ions. This would result in no net change in the oxidation state of the bromine species present in the system. If reduction of water is to occur in the presence of bromine the ^{-ve} electrode in the ^{-ve} chamber must be specially constructed to starve it of bromine. Inclusion of such specially constructed electrodes is clearly undesirable from an economic and system maintenance viewpoint.

The rebalancing process may be applied continuously to the RFC wherein a sidestream of the bromine/bromide or sulfide/polysulfide electrolyte drawn from the mainstream is diverted through apparatus suitable for carrying out the rebalancing process. The rebalancing process may also be applied as a batch process wherein the fraction of the bromine/bromide or sulfide/polysulfide electrolyte which is removed from the RFC is treated in separate apparatus suitable for carrying out the rebalancing process before being returned to the RFC.

It will be understood by those skilled in the art that a number of reduction half-cell reactions may be used to counter the oxidation of the halide or sulfide. However, in the present invention, the other half-cell reaction under alkaline conditions involves the reduction of water to hydrogen and hydroxide ions according to the half-cell reaction shown in Equation 8 below:

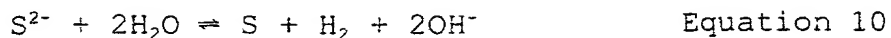
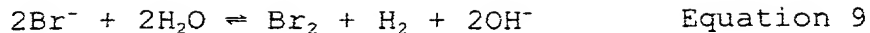


Equation 8

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Thus the rebalancing process may be represented by the reactions shown in Equations 9 and 10 below:

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5 Similarly in an acidic medium the half cell reaction comprises



10 It will be appreciated that, although the process of oxidising the bromine/bromide or sulfide/polysulfide electrolyte can be used to rebalance the electrolytes, there is still a net loss of active sulfur species from the cell. This is because the sulfide and
15 polysulfide ions which cross to the bromine electrolyte and are oxidised to sulfate ions are not recovered. Thus, in a preferred embodiment of the present invention, the process additionally comprises adding elemental sulfur or a sulfide salt to the
20 sulfide/polysulfide electrolyte in an amount such as to restore the initial concentration of active sulfur species.

25 In carrying out the process of the present invention the electrolyte circulating through the -^{ve} chamber of the auxiliary cell may be water. In this instance the electrolyte will generally circulate in a closed system and there will be no change of pH of the bromine/bromide or sulfide/polysulfide electrolyte.

30 In an alternative manner of carrying out the process of the present invention the electrolyte circulating through the -^{ve} chamber of the auxiliary cell is a fraction of electrolyte 1 or 2 which has been made
35 free of polysulfide or bromine by electrochemical reduction thereof. This may be achieved by recirculating electrolyte 1 or 2 through the -^{ve}

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chamber of the auxiliary cell until all of the polysulfide or bromine has been reduced. The electrolyte circulating through the $-ve$ chamber of the auxiliary cell may then be returned to the main stream of electrolyte 1 or 2.

Alternatively, the electrochemical reduction of polysulfide or bromine which may be present in electrolyte 1 or 2 respectively occurs within the $-ve$ chamber of a second auxiliary cell which comprises a $+ve$ chamber containing an inert $+ve$ electrode and a $-ve$ chamber containing an inert $-ve$ electrode, the chambers being separated from one another by a cation exchange membrane, the electrolyte circulating through the $+ve$ chamber being a fraction of electrolyte 1 or electrolyte 2. This may be achieved by recirculating electrolyte 1 or 2 through the $-ve$ chamber of the second auxiliary cell until all of the polysulfide or bromine has been reduced. The electrolyte circulating through the $-ve$ chamber of the auxiliary cell may then be returned to the main stream of electrolyte 1 or 2.

Another reason why the reduction of any bromine which may be present in electrolyte 2 is important is because, as described in WO-A-00/03448, carrying out the RFC process of the present invention results in the production of sulfate ions in the bromine/bromide electrolyte as described above with reference to Equation 7. The removal of sulfate ions from the electrolyte can only be carried out by the process as described in WO-A-00/03448 in the absence of free bromine which otherwise interferes with the process. Thus, in a preferred embodiment, the electrolyte circulating through the $-ve$ chamber of the auxiliary cell during rebalancing is a fraction of electrolyte 2 and that fraction is subsequently treated to remove sulfate ions contained therein.

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In carrying out the process of the present invention elemental sulfur and/or a sulfide salt may be added to the sulfide/polysulfide electrolyte in an amount sufficient to restore the initial concentration of sulfur species.

The present invention also provides for the use, in a process for energy storage and/or power delivery comprising:

- 10 (i) maintaining and circulating electrolyte flows in a fully liquid system in which the active constituents are fully soluble in a single cell or in an array of repeating cell structures, each cell with a positive (+^{ve}) chamber containing an
15 inert +^{ve} electrode and a negative (-^{ve}) chamber containing an inert -^{ve} electrode, the chambers being separated from one another by a cation exchange membrane, the electrolyte circulating in the -^{ve} chamber of each cell during power
20 delivery containing a sulfide (electrolyte 1), and the electrolyte circulating in the +^{ve} chamber during power delivery containing bromine (electrolyte 2),
- 25 (ii) restoring or replenishing the electrolytes in the +^{ve} and -^{ve} chambers by circulating the electrolyte from each chamber to storage means comprising a volume of electrolyte greater than the cell volume for extended delivery of power over a longer discharge cycle than the cell
30 volume alone would permit,
of a process comprising:
circulating a fraction of electrolyte 1 or electrolyte 2 through the +^{ve} chamber of an auxiliary cell, said auxiliary cell comprising a
35 +^{ve} chamber containing an inert +^{ve} electrode and a -^{ve} chamber containing an inert -^{ve} electrode, the chambers being separated from one another by

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a cation exchange membrane, the electrolyte circulating through the $-ve$ chamber of the auxiliary cell containing water and being free from polysulfide and free from bromine during rebalancing, the auxiliary cell operating so as to oxidise sulfide ions to polysulfide or bromide ions to bromine in the $+ve$ chamber and so as to reduce water to hydrogen and hydroxide ions in the $-ve$ chamber,

for the purpose of rebalancing electrolytes 1 and 2.

The present invention also includes within its scope apparatus for carrying out a process as described above comprising:

- (i) a single cell or an array of repeating cell structures, each cell comprising; a $+ve$ chamber containing an inert $+ve$ electrode and a $-ve$ chamber containing an inert $-ve$ electrode the chambers being separated from one another by an ion exchange membrane, an electrolyte circulating in the $-ve$ chamber of each cell which contains a sulfide during power delivery (electrolyte 1), and an electrolyte circulating in the $+ve$ chamber which contains bromine during power delivery (electrolyte 2),
- (ii) storage and circulation means for each electrolyte for restoring or replenishing the electrolytes in the $+ve$ and $-ve$ chambers,
- (iii) means for rebalancing the electrolytes comprising an auxiliary cell which comprises a $+ve$ chamber containing an inert $+ve$ electrode and a $-ve$ chamber containing an inert $-ve$ electrode the chambers being separated from one another by a cation exchange membrane, means for circulating a fraction of electrolyte 1 or 2 through the $+ve$ chamber of the auxiliary cell, an electrolyte containing water and being free from polysulfide

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and free from bromine during rebalancing and means for circulating said electrolyte through the -ve chamber of the auxiliary cell.

5 The present invention will be further described with reference to the accompanying drawings in which:

Fig 1A is a schematic view of a basic electrochemical reduction-oxidation cell in which a
10 sulfide/polysulfide reaction is carried out in one half of the cell and a bromine/bromide reaction is carried out in the other half of the cell;

Fig 1B is a diagram of cell arrays using the system of
15 Fig. 1A;

Fig 2 is a block diagram of a fluid flow system using the cell of Fig 1A;

20 Fig 3 is a flow diagram of an apparatus for carrying out a preferred embodiment of the process of the present invention.

Fig 4 is a flow diagram of an apparatus for carrying
25 out a preferred embodiment of the process of the present invention.

Fig 5 is a schematic diagram of an apparatus for carrying out a further preferred embodiment of the
30 process of the present invention, including the removal of sulfate.

Fig 6 is a flow diagram of an apparatus for carrying
35 out a preferred embodiment of the process of the present invention, including the removal of sulfate.

Fig 7 is a graph of voltage versus time for a selected

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number of cycles of a RFC which does not incorporate a rebalancing process in accordance with the present invention.

5 Fig 8 is a graph of voltage versus time for a selected number of cycles of a RFC which does incorporate a rebalancing process in accordance with the present invention.

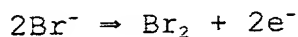
10 Fig 1A shows a cell 10 with a positive (+^{ve}) electrode 12 and a negative (-^{ve}) electrode 14 and a cation exchange membrane 16 which may be formed from a fluorocarbon polymer with sulfonic acid functional groups to provide charge carriers. The membrane 16
15 acts to separate the +^{ve} and -^{ve} sides of the cell 10 and is selected to minimize migration of bromine from the +^{ve} side to the -^{ve} side and to minimize migration of sulfide and polysulfide ions from the -^{ve} side to the +^{ve} side. An aqueous solution 22 of NaBr is
20 provided in a chamber 22C formed between the +^{ve} electrode 12 and the membrane 16 and an aqueous solution 24 of Na₂S_x (where x may be from 2 to 5) is provided in a chamber 24C formed between the -^{ve} electrode 14 and the membrane 16. A K₂S_x solution,
25 which is more soluble and more expensive than the Na₂S_x solutions, is used in another embodiment.

When the cell is in the discharged state, a solution of NaBr of up to 6.0 molar concentration
30 exists in the chamber 22C of the cell and a solution of Na₂S_x at 0.5 to 1.5 molar, exists in chamber 24C of the cell. Higher molarity is possible with K₂S_x.

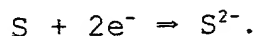
As the cell is charged, Na⁺ ions are transported
35 through the cation membrane 16, as shown in Fig 1A, from the +^{ve} to the -^{ve} side of the cell. Free bromine is produced via oxidation of the bromide ions at the

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+^{ve} electrode and dissolves as a tribromide or pentabromide ion. Sulfur is reduced at the -^{ve} electrode and the pentasulfide, Na₂S_x, salt eventually becomes the monosulfide as the charging proceeds to completion. At the +^{ve} side the following reaction occurs,



and at the -^{ve} side the following reaction occurs,



The membrane separates the two electrolytes and prevents bulk mixing and also retards the migration of sulfide and polysulfide ions from the -^{ve} side to the +^{ve} side, and the migration of Br⁻ and Br₂ from the +^{ve} to the -^{ve} side. Diffusion of the sulfide and polysulfide ions across the membrane results in the electrolytes becoming unbalanced as described earlier.

When providing power, the cell is discharging. During this action, reversible reactions occur at the two electrodes. At the +^{ve} side electrode 12, bromine is reduced to Br⁻, and at the -^{ve} electrode, the S²⁻ ion is oxidized to molecular S. The electrons produced at the -^{ve} electrode form the current through a load. The chemical reaction at the +^{ve} electrode produces 1.06 to 1.09 volts and the chemical reaction at the -^{ve} electrode produces 0.48 to 0.52 volts. The combined chemical reactions produce an open circuit voltage of 1.54 to 1.61 volts per cell.

The present system is an anionically active electrochemical system. Therefore, the cation which is associated with them essentially takes no part in the energy producing process. Hence, a cation of

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"convenience" is chosen. Sodium or potassium are preferred choices. Sodium and potassium compounds are plentiful, they are inexpensive and have high water solubilities. Lithium and ammonium salts are also possibilities, but at higher costs.

Fig 1B shows an array 20 of multiple cells connected in electrical series and fluid parallel. Multiple mid-electrodes 13 (each one having a +^{ve} electrode side 12A and -^{ve} electrode side 14A) and end electrodes 12E (+^{ve}) and 14E (-^{ve}) are spaced out from each other by membranes 16 and screen or mesh spacers (22D, 24D) in all the cell chambers 22C, 24C, (portions of two of which 22D, 24D are shown by way of example) to form end cells C_{E1} and C_{E2} and an array of mid cells C_M (typically 10-20; but note much smaller and much higher numbers of cells can be accommodated). The end electrodes 12E (+^{ve}) and 14E (-^{ve}) have internal conductors 12F and 14F (typically copper screens) encapsulated therein and leading to external terminals 12G, 14G which are connected to external loads (e.g. to motor(s) via a control circuit (CONT), the motor(s) may be used to drive a vehicle) or power sources (e.g. utility power grid when used as a load-levelling device).

Fig 2 shows a free flow system, a power generation/storage system utilizing one or more of the batteries or cell array formats 20. Each cell 20 receives electrolyte through pumps 26 and 28 for the NaBr and Na₂S_x solutions (22 and 24, respectively). The electrolytes 22 and 24 are stored in containers 32 and 34. The tanks 32, 34 can be replaced with freshly charged electrolyte by substituting tanks containing fresh electrolyte and/or refilling them from charged supply sources via lines 32R, 34R with corresponding lines (not shown) provided for draining spent

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(discharged) reagent. The electrolytes 22 and 24 are pumped from tanks 32 and 34, respectively, into the respective chambers 22C and 24C by means of pumps 26 and 28.

5

Fig 3 shows a free flow system in which an array of cells 20 are supplied with bromine/bromide and sulfide/polysulfide electrolyte from storage tanks 41 and 42 via lines 43 and 44. Bromine/bromide electrolyte may be removed from storage tank 41 via line 50 which transfers it to an external electrochemical cell 51 wherein the bromide is oxidised to bromine as a half-cell reaction in an electrochemical process. The other half-cell reaction involves the reduction of water to hydrogen and hydroxide ions. An aqueous electrolyte is stored in tank 52 and transported to the electrochemical cell 51 via line 53. The reduced electrolyte is passed via line 54 to tank 55 where hydrogen gas which is generated by the electrochemical reaction may be vented from the system. The electrolyte returns via line 56 to storage tank 52.

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Fig 4 shows a particularly preferred variation of the free flow system illustrated in Fig 3. In this embodiment the aqueous electrolyte which is reduced in the external electrochemical cell 51 is also bromine/bromide electrolyte which has been removed from storage tank 41 to tank 52 via line 57. In this case, the reduction reaction will initially involve reduction of any residual bromine to bromide and will subsequently involve reduction of water to hydrogen and hydroxide ions. The reduced electrolyte may be subsequently returned to the storage tank 41 along line 58.

Referring to Fig 5, a schematic flow diagram is shown

Fig 6 shows an alternative system for use in accordance with the present invention which includes a sulfate crystallization unit as described in WO-A-00/03448. In this system a portion of bromide/bromine

Fig 6 shows an alternative system for use in accordance with the present invention which includes a sulfate crystallization unit as described in WO-A-00/03448. In this system a portion of bromide/bromine

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electrolyte contaminated with sulfate ions is drawn from the main system 80 via line 81 and held in a receiving tank 82. This electrolyte is then circulated via lines 83 and 84 through the -^{ve} chamber 85 of an auxiliary cell 86 until substantially all of the bromine present in the electrolyte has been reduced to bromide ions. The voltage applied across cell 86 is limited to ensure that reduction of water does not occur. When the current density has run down (thus indicating that the conversion of bromine to bromide is substantially complete), the voltage applied to the cell 86 is increased to a value sufficient to cause reduction of the water present in the electrolyte so as to generate H₂ gas and OH⁻ ions. The electrolyte circulating through the +^{ve} chamber of the external auxiliary cell is either sulfide/polysulfide or bromine/bromide electrolyte taken from the main system. The oxidation of one or the other of these electrolytes rebalances the system. The removal of water from the electrolyte is advantageous because it further increases the concentration of bromide ions thereby reducing the sulfate solubility and increasing the yield of sulfate on crystallisation. A tap 87 is provided to draw off H₂ gas. When sufficient rebalancing has occurred, the electrolyte circulating through the -^{ve} chamber 85 is passed from the receiving tank 82 to the crystalliser 88 via line 89. This electrolyte is then passed via line 90 through a filter 91 to remove the sulfate crystals and then it may be returned to the main system via line 92. The embodiment shown in Fig 6 shows an electrolyte being drawn from the main system via line 93, passing through the +^{ve} chamber 94 of the auxiliary cell and returning the main system via line 95.

The present invention will now be further described by

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reference to the following examples.

Comparative Example 1

5 A regenerative fuel cell of the type described above having sulfide/polysulfide and bromine/bromide electrolytes was set up. The cell had the following specifications:

10 electrode material: polyethylene impregnated with activated carbon
electrode area: 2000cm²
current density: 80mA/cm²
electrolyte volume: 9l per electrolyte
15 cycle time: 6hours (i.e. 3hours charge and 3hours discharge)
flow rate: 1000ml/min
membrane material: Nafion 115™

20 The cell was operated over 18 cycles (108 hours) and the cell voltage was monitored throughout this period. The results are shown in Fig 7. It can be seen that after a limited number of cycles (about 7) the cell voltage limits early on the discharge cycle due to the lack of bromine. This problem gets worse as the
25 number of cycles increases. It can be clearly seen on the graph that after about 12 cycles (72 hours) the cell fails to maintain a good voltage performance over the whole of the 3 hour discharge cycle.

30 Example 1

An identical RFC to that used in Comparative Example 1 was set up. This time the electrolytes were continuously rebalanced by oxidation of a sidestream of the bromide/bromine electrolyte drawn from the
35 mainstream. Oxidation occurred in one half of an external electrochemical cell wherein the electrolyte

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undergoing reduction in the other half of the cell was dilute aqueous sodium hydroxide. The external electrochemical cell used was an MP cell from Electrocell AB having the following specifications:

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anode material: platinum
cathode material: nickel
electrode area: 100cm²
current density: 13mA/cm²
10 flow rate: 270ml/min
membrane material: Nafion 350™

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Fig.3 shows a schematic representation of the apparatus used in the present example. The cell was operated over at least 92 cycles (552 hours) and the cell voltage was monitored throughout this period. The results from the period from 400 to 550 hours are shown in Fig 8. It can be seen that even after 91 cycles (546 hours) the cell voltage does not limit early on the discharge cycle as occurred in the unbalanced cell. The cell retains a good voltage performance over the whole of the 3 hour discharge cycle.

CLAIMS:

1. An electrochemical process for energy storage and/or power delivery comprising:
- 5 (i) maintaining and circulating electrolyte flows in a fully liquid system in which the active constituents are fully soluble in a single cell or in an array of repeating cell structures, each cell with a positive (+^{ve}) chamber containing an
10 inert +^{ve} electrode and a negative (-^{ve}) chamber containing an inert -^{ve} electrode, the chambers being separated from one another by a cation exchange membrane, the electrolyte circulating in the -^{ve} chamber of each cell during power
15 delivery containing a sulfide (electrolyte 1), and the electrolyte circulating in the +^{ve} chamber during power delivery containing bromine (electrolyte 2),
- (ii) restoring or replenishing the electrolytes in the
20 +^{ve} and -^{ve} chambers by circulating the electrolyte from each chamber to storage means comprising a volume of electrolyte greater than the cell volume for extended delivery of power over a longer discharge cycle than the cell
25 volume alone would permit, and
- (iii) rebalancing the electrolytes by circulating a fraction of electrolyte 1 or electrolyte 2 through the +^{ve} chamber of an auxiliary cell, said auxiliary cell comprising a +^{ve} chamber
30 containing an inert +^{ve} electrode and a -^{ve} chamber containing an inert -^{ve} electrode, the chambers being separated from one another by a cation exchange membrane, the electrolyte circulating through the -^{ve} chamber of the
35 auxiliary cell containing water and being free from polysulfide and free from bromine during rebalancing, the auxiliary cell operating so as

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to oxidise sulfide ions to sulfur or bromide ions to bromine in the +^{ve} chamber and so as to reduce water to hydrogen and hydroxide ions in the -^{ve} chamber.

5

2. A process as claimed in claim 1 wherein the electrolyte circulating through the -^{ve} chamber of the auxiliary cell during rebalancing is a fraction of electrolyte 1 or electrolyte 2 which has been made
10 free of polysulfide or bromine by electrochemical reduction thereof.

3. A process as claimed in claim 2 wherein the electrochemical reduction of polysulfide or bromine is
15 effected by recirculating the fraction of electrolyte 1 or 2 through the -^{ve} chamber of the auxiliary cell until all of the polysulfide or bromine has been reduced.

20 4. A process as claimed in claim 2 wherein the electrochemical reduction of polysulfide or bromine occurs within the -^{ve} chamber of a second auxiliary cell which comprises a +^{ve} chamber containing an inert +^{ve} electrode and a -^{ve} chamber containing an inert -^{ve}
25 electrode, the chambers being separated from one another by a cation exchange membrane, the electrolyte circulating through the +^{ve} chamber being a fraction of electrolyte 1 or electrolyte 2.

30 5. A process as claimed in claim 4 wherein the electrochemical reduction of polysulfide or bromine is effected by recirculating the fraction of electrolyte 1 or 2 through the -^{ve} chamber of the second auxiliary cell until all of the polysulfide or bromine has been
35 reduced.

6. A process as claimed in any one of claims 3 to 5

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wherein the electrolyte circulating through the $-ve$ chamber of the auxiliary cell during rebalancing is a fraction of electrolyte 2 and wherein that fraction is subsequently treated to remove sulfate ions contained therein.

7. A process as claimed in claim 6 wherein said sulfate ions are removed by crystallisation of a sulfate salt from the fraction of electrolyte 2.

8. A process as claimed in any one of claims 2 to 7 wherein the fraction of electrolyte 1 or 2 which is circulated through the $-ve$ chamber of the auxiliary cell is returned to the main stream of electrolyte 1 or 2 respectively.

9. A process as claimed in any one of the preceding claims which additionally comprises adding elemental sulfur and/or a sulfide salt to electrolyte 1 in an amount sufficient to restore the initial concentration of sulfur species.

10. Use, in a process for energy storage and/or power delivery comprising:

(i) maintaining and circulating electrolyte flows in a fully liquid system in which the active constituents are fully soluble in a single cell or in an array of repeating cell structures, each cell with a positive ($+ve$) chamber containing an inert $+ve$ electrode and a negative ($-ve$) chamber containing an inert $-ve$ electrode, the chambers being separated from one another by a cation exchange membrane, the electrolyte circulating in the $-ve$ chamber of each cell during power delivery containing a sulfide (electrolyte 1), and the electrolyte circulating in the $+ve$ chamber during power delivery containing bromine

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(electrolyte 2),
(ii) restoring or replenishing the electrolytes in the
+^{ve} and -^{ve} chambers by circulating the
electrolyte from each chamber to storage means
5 comprising a volume of electrolyte greater than
the cell volume for extended delivery of power
over a longer discharge cycle than the cell
volume alone would permit,

of a process comprising:

10 circulating a fraction of electrolyte 1 or
electrolyte 2 through the +^{ve} chamber of an
auxiliary cell, said auxiliary cell comprising a
+^{ve} chamber containing an inert +^{ve} electrode and
a -^{ve} chamber containing an inert -^{ve} electrode,
15 the chambers being separated from one another by
a cation exchange membrane, the electrolyte
circulating through the -^{ve} chamber of the
auxiliary cell containing water and being free
from polysulfide and free from bromine during
20 rebalancing, the auxiliary cell operating so as
to oxidise sulfide ions to polysulfide or bromide
ions to bromine in the +^{ve} chamber and so as to
reduce water to hydrogen and hydroxide ions in
the -^{ve} chamber,

25 for the purpose of rebalancing electrolytes 1 and 2.

11. An electrochemical apparatus for energy storage
and/or power delivery comprising:

(i) a single cell or an array of repeating cell
30 structures, each cell comprising; a +^{ve} chamber
containing an inert +^{ve} electrode and a -^{ve}
chamber containing an inert -^{ve} electrode the
chambers being separated from one another by an
ion exchange membrane, an electrolyte circulating
35 in the -^{ve} chamber of each cell which contains a
sulfide during power delivery (electrolyte 1),
and an electrolyte circulating in the +^{ve} chamber

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which contains bromine during power delivery
(electrolyte 2),

- (ii) storage and circulation means for each
electrolyte for restoring or replenishing the
5 electrolytes in the +^{ve} and -^{ve} chambers,
(iii) means for rebalancing the electrolytes
comprising an auxiliary cell which comprises a
+^{ve} chamber containing an inert +^{ve} electrode and
a -^{ve} chamber containing an inert -^{ve} electrode
10 the chambers being separated from one another by
a cation exchange membrane, means for circulating
a fraction of electrolyte 1 or 2 through the +^{ve}
chamber of the auxiliary cell, an electrolyte
containing water and being free from polysulfide
15 and free from bromine during rebalancing and
means for circulating said electrolyte through
the -^{ve} chamber of the auxiliary cell.

12. Apparatus as claimed in claim 11 wherein the
20 means for circulating an electrolyte through the -^{ve}
chamber of the auxiliary cell comprises means for
circulating a fraction of electrolyte 1 or 2 through
the -^{ve} chamber of the auxiliary cell.

- 25 13. Apparatus as claimed in claim 11 wherein the
means for circulating an electrolyte through the -^{ve}
chamber of the auxiliary cell comprises a storage tank
into which a fraction of electrolyte 1 or 2 may be
transferred and means for re-circulating the fraction
30 of electrolyte 1 or 2 between the -^{ve} chamber of the
auxiliary cell and said storage tank.

14. Apparatus as claimed in claim 12 or claim 13
which additionally comprises a second auxiliary cell
35 which comprises a +^{ve} chamber containing an inert +^{ve}
electrode and a -^{ve} chamber containing an inert -^{ve}
electrode, the chambers being separated from one

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another by a cation exchange membrane, means for circulating a fraction of electrolyte 1 or 2 through the +^{ve} chamber and means for circulating a fraction of electrolyte 1 or 2 through the -^{ve} chamber.

5

15. Apparatus as claimed in claim 14 wherein the means for circulating an electrolyte through the -^{ve} chamber of the second auxiliary cell comprises a storage tank into which a fraction of electrolyte 1 or 2 may be transferred and means for re-circulating the fraction of electrolyte 1 or 2 between the -^{ve} chamber of the second auxiliary cell and said storage tank.

10

16. Apparatus as claimed in any one of claims 12 to 15 wherein the electrolyte circulated through the -^{ve} chamber of the auxiliary cell is electrolyte 2, additionally comprising means for removing sulfate ions from the fraction of electrolyte 2 after circulation through the -^{ve} chamber of the auxiliary cell.

15

20

17. Apparatus as claimed in claim 16 wherein the means for removing sulfate ions from electrolyte 2 comprises a crystalliser.

25

18. Apparatus as claimed in any one of claims 11 to 17 additionally comprising means for passing the fraction of electrolyte 1 or 2 which is circulated through the -^{ve} chamber of the auxiliary cell back to the main stream of electrolyte 1 or 2 respectively.

30

19. Use in an electrochemical apparatus for energy storage and/or power delivery comprising:

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- (i) a single cell or an array of repeating cell structures, each cell comprising; a +^{ve} chamber containing an inert +^{ve} electrode and a -^{ve} chamber containing an inert -^{ve} electrode the

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chambers being separated from one another by an ion exchange membrane, an electrolyte circulating in the $-ve$ chamber of each cell which contains a sulfide during power delivery (electrolyte 1),
5 and an electrolyte circulating in the $+ve$ chamber which contains bromine during power delivery (electrolyte 2), and

(ii) storage and circulation means for each electrolyte for restoring or replenishing the
10 electrolytes in the $+ve$ and $-ve$ chambers,

01

an auxiliary cell which comprises; a $+ve$ chamber containing an inert $+ve$ electrode and a $-ve$
15 chamber containing an inert $-ve$ electrode the chambers being separated from one another by a cation exchange membrane, means for passing a fraction of electrolyte 1 or 2 through the $+ve$ chamber of the auxiliary cell, an electrolyte
20 containing water and being free from polysulfide and free from bromine during rebalancing and means for circulating said electrolyte through the $-ve$ chamber of the auxiliary cell

for the purpose of rebalancing electrolytes 1 and 2.

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FIG. 1A.

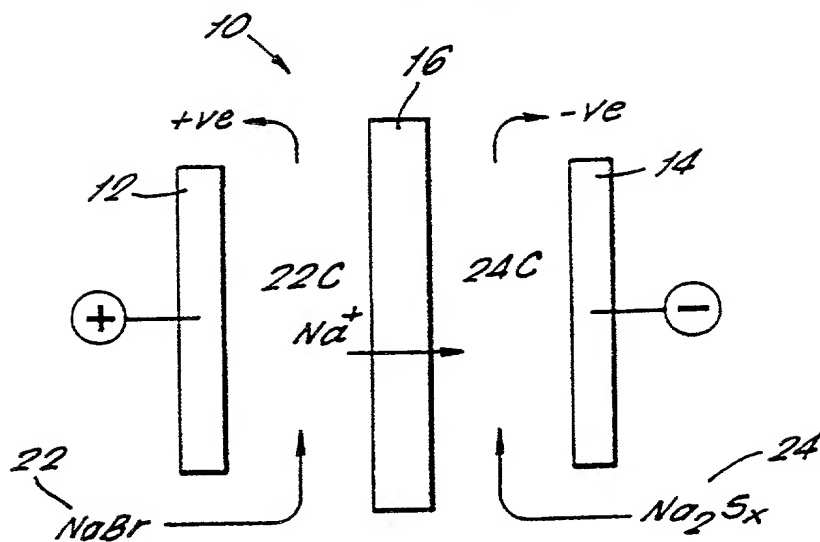
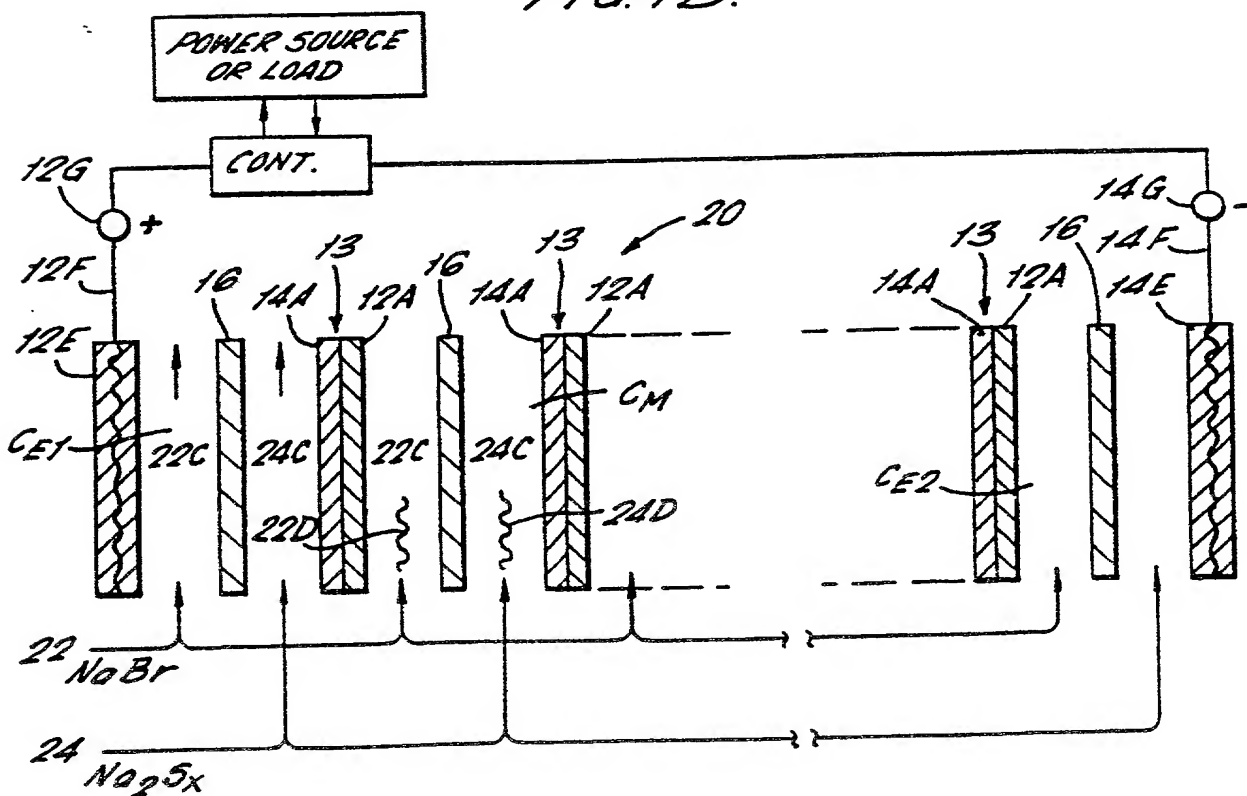


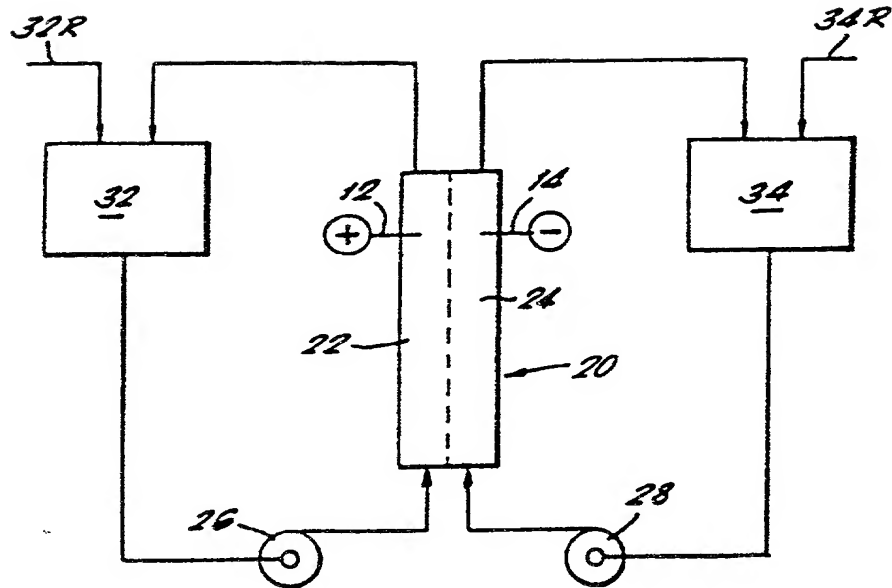
FIG. 1B.



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FIG. 2.

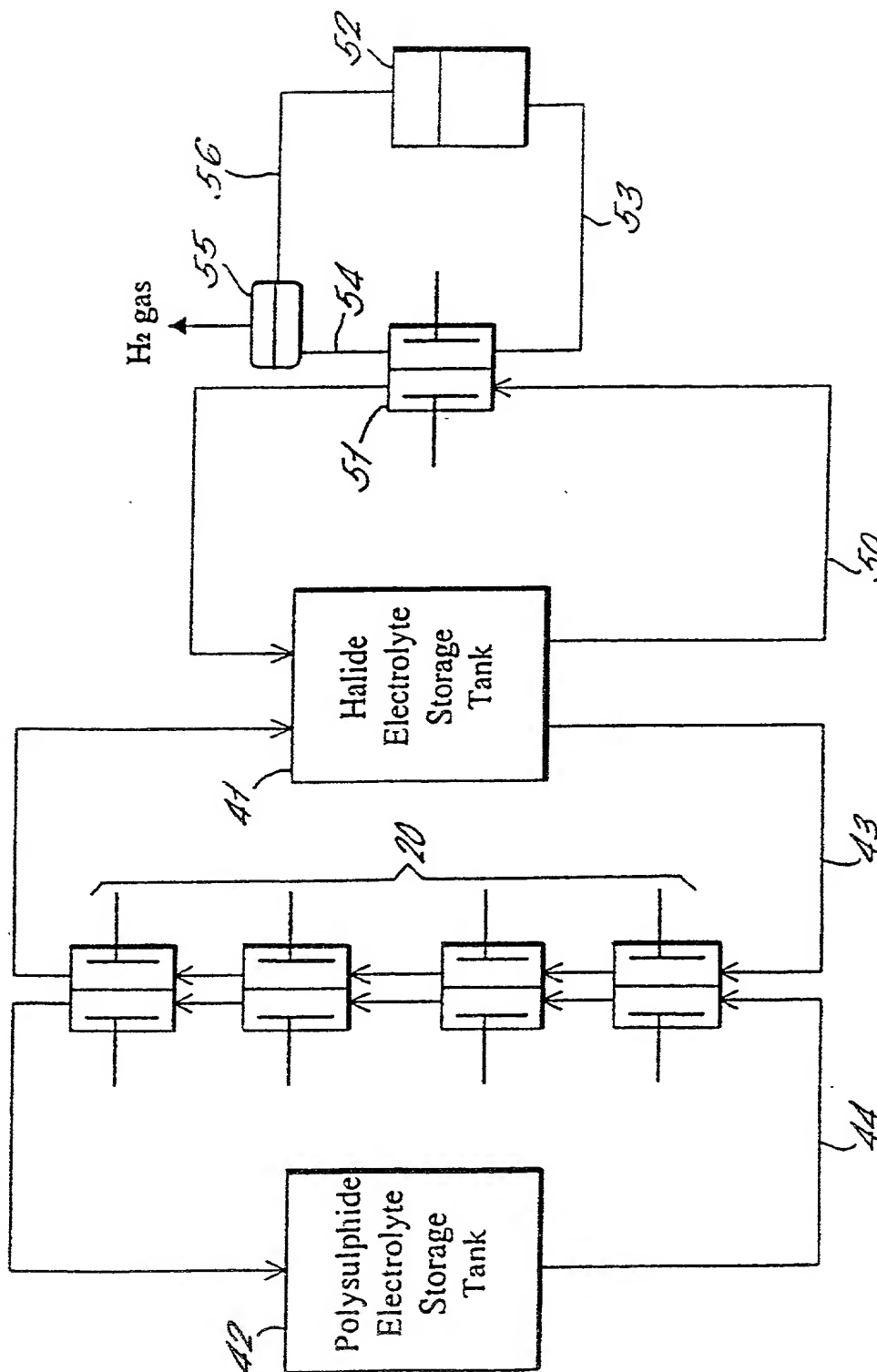


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FIG. 3.



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FIG. 4.

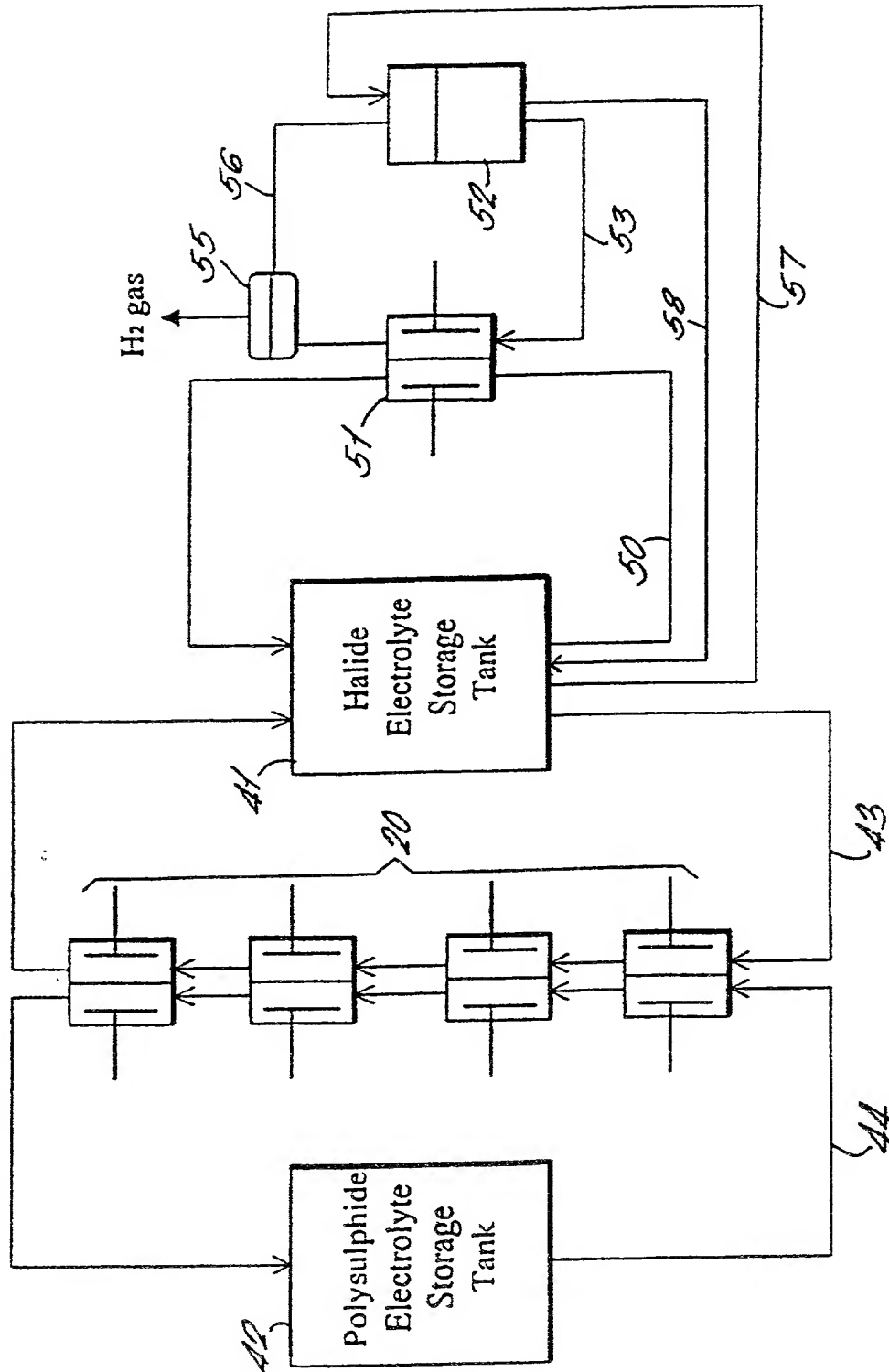
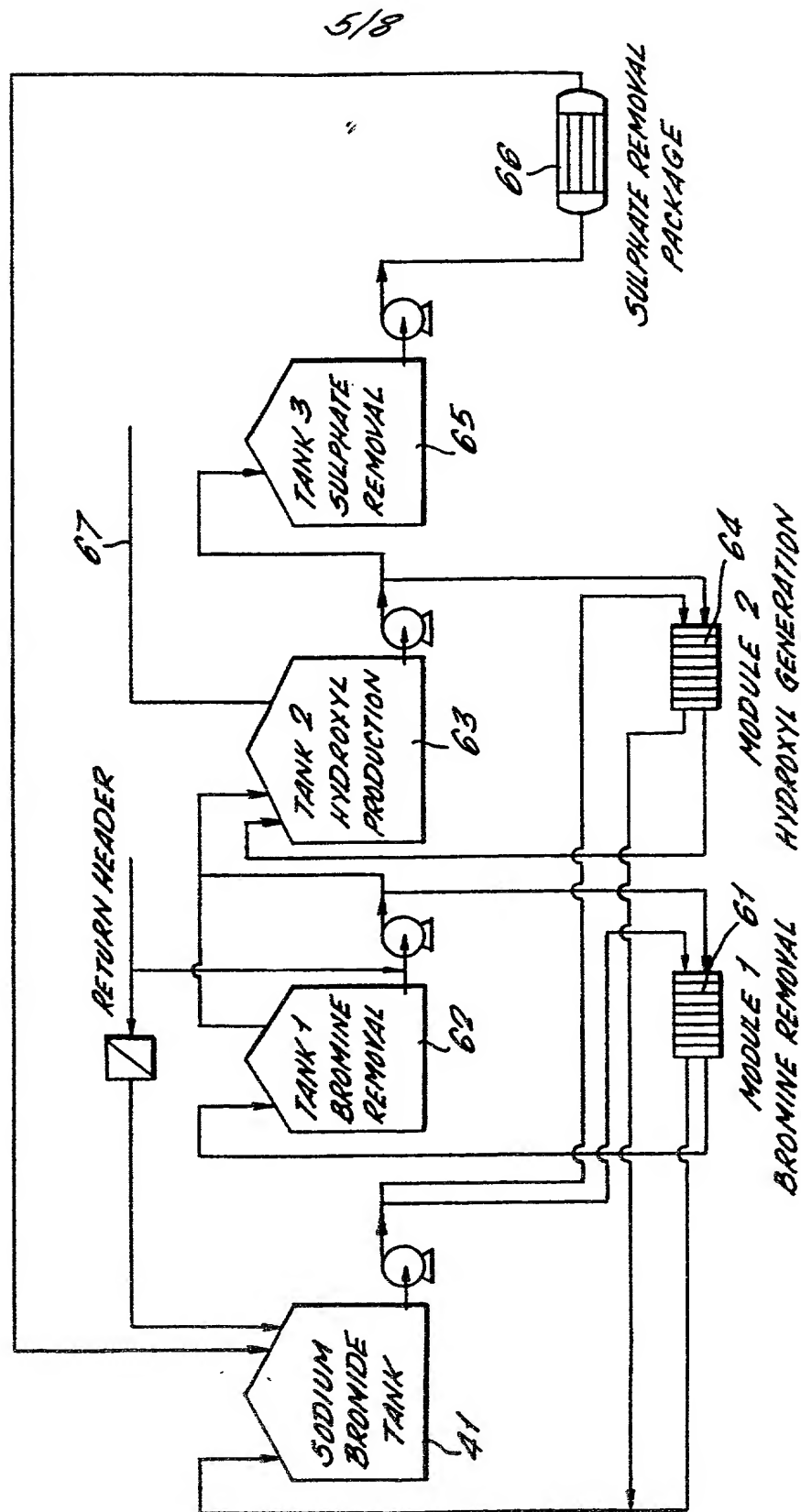
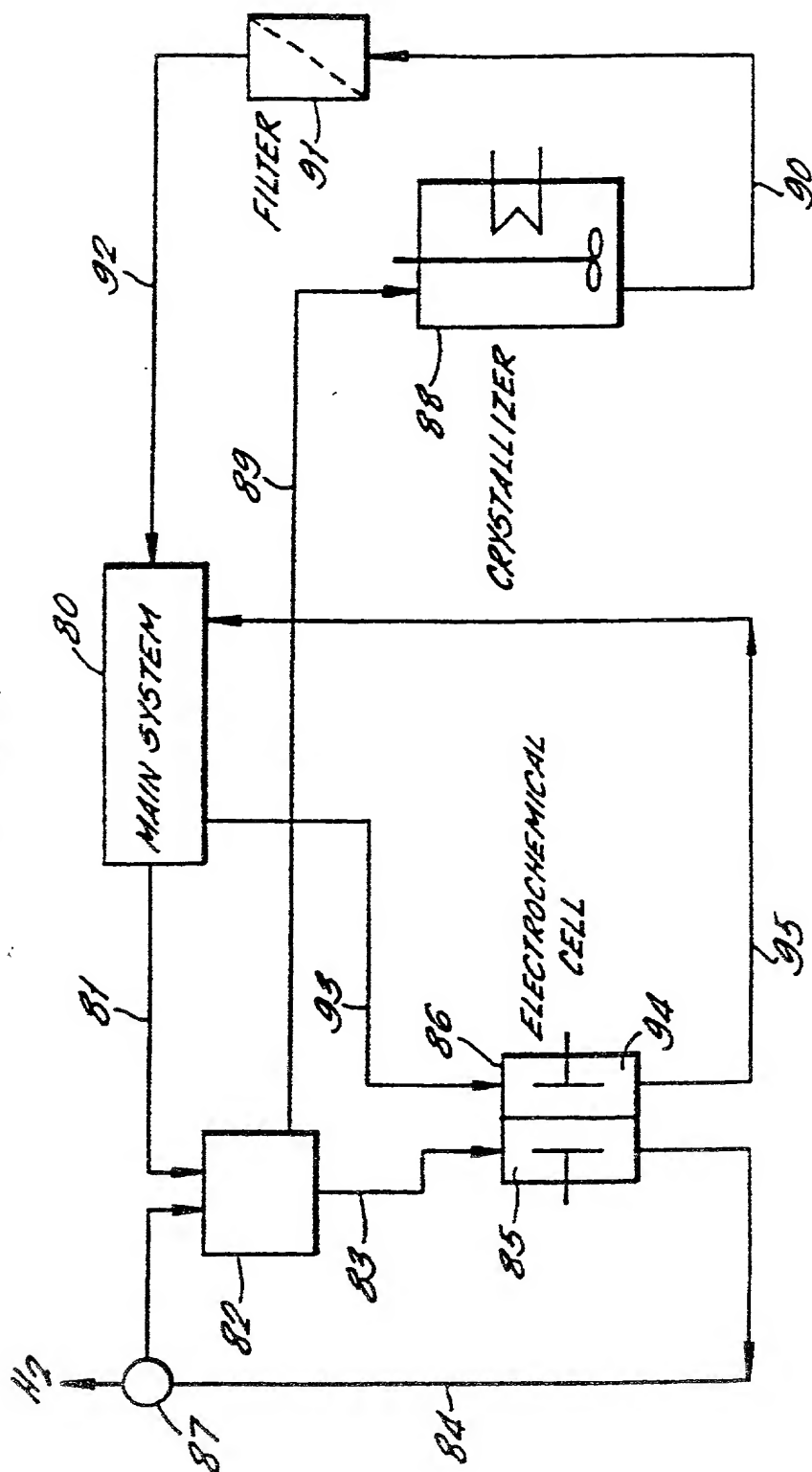


FIG. 5.



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FIG. 6.



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FIG. 7.

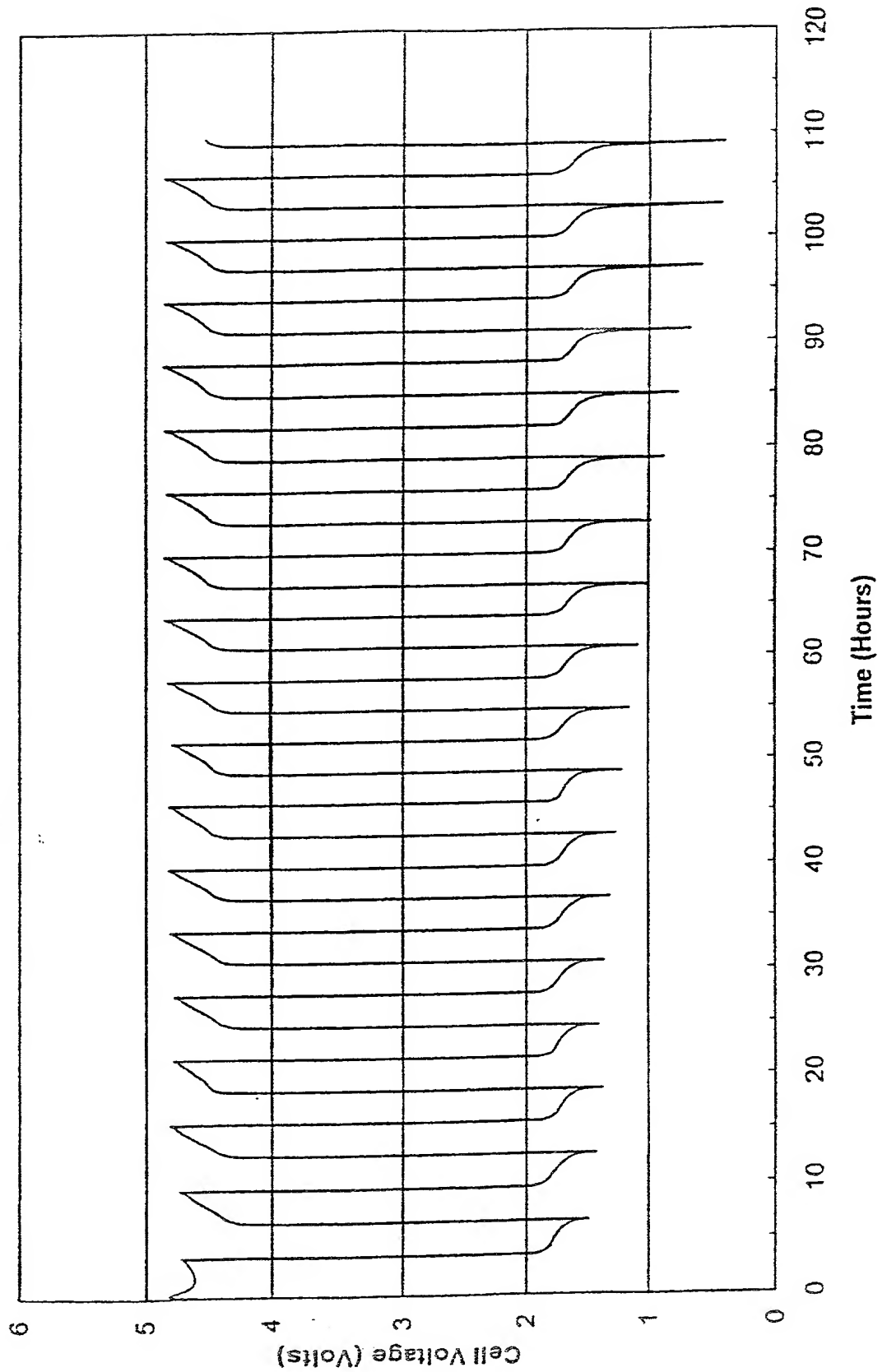
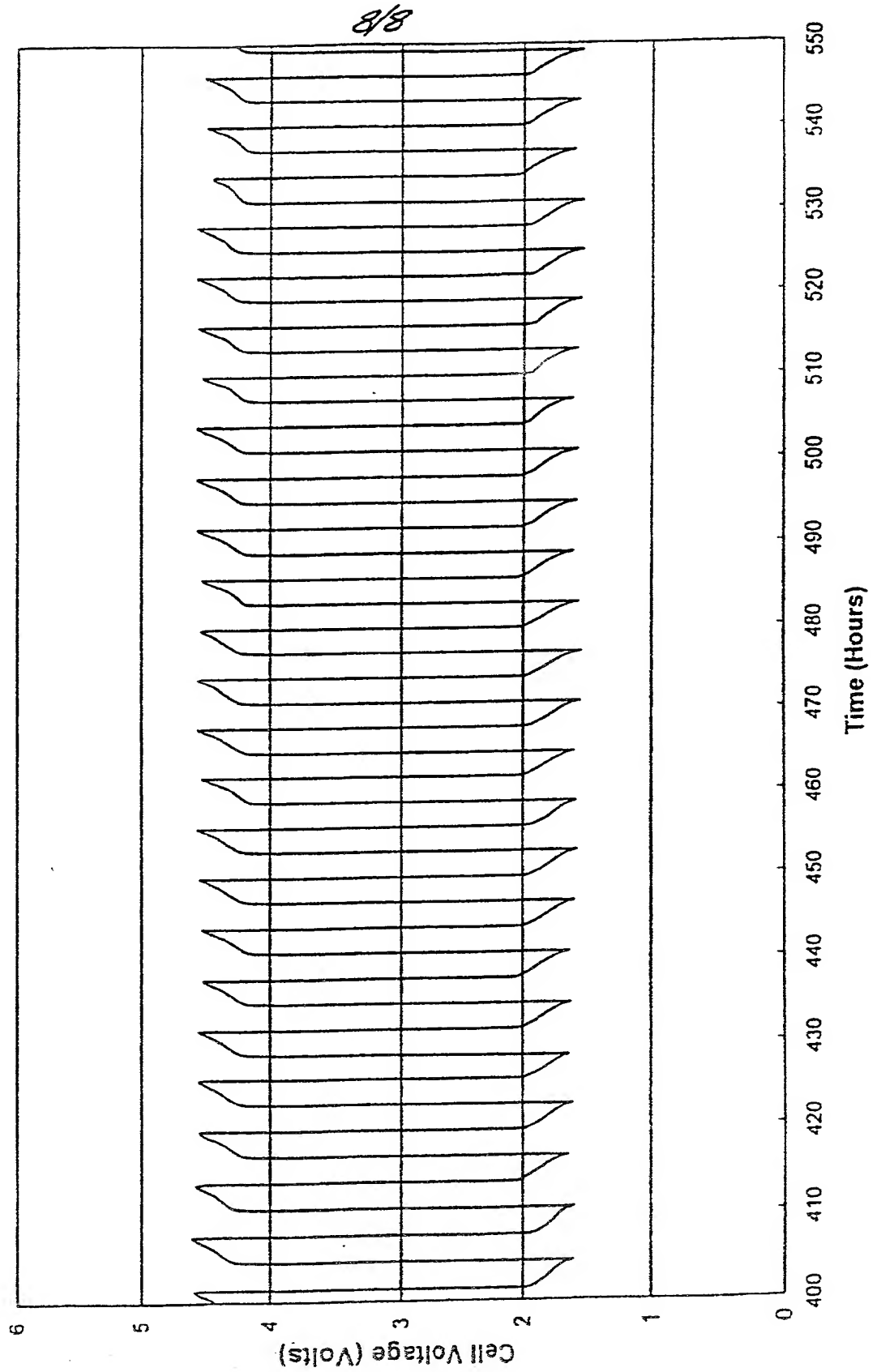


FIG. 8.



DECLARATION AND POWER OF ATTORNEY FILED WITH U.S. DESIGNATED OFFICE UNDER 35 U.S.C. 371(c)(4)

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name, I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

ELECTROLYTE REBALANCING SYSTEM

the specification of which was filed as PCT International Application No. PCT/GB00/02536
filed 30th June, 2000 and was amended on _____ (if applicable)

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, § 1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, § 119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)	Priority Claimed
<u>PCT/GB00/02103</u> (Number)	<u>GB</u> (Country)
<u>9928344.2</u> (Number)	<u>GB</u> (Country)
<u>2/7/1999</u> (Day/Month/Year Filed)	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
<u>30/11/1999</u> (Day/Month/Year Filed)	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
<u> </u> (Day/Month/Year Filed)	<input type="checkbox"/> Yes <input type="checkbox"/> No
<u> </u> (Day/Month/Year Filed)	<input type="checkbox"/> Yes <input type="checkbox"/> No
<u> </u> (Day/Month/Year Filed)	<input type="checkbox"/> Yes <input type="checkbox"/> No
<u> </u> (Day/Month/Year Filed)	<input type="checkbox"/> Yes <input type="checkbox"/> No

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, § 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)	(Filing Date)	(Status: patented, pending, abandoned)
<u> </u>	<u> </u>	<u> </u>
<u> </u>	<u> </u>	<u> </u>
<u> </u>	<u> </u>	<u> </u>
<u> </u>	<u> </u>	<u> </u>

I hereby appoint as principal attorneys: Donald R. Antonelli, Reg. No. 20,296; David T. Terry, Reg. No. 20,178; Melvin Kraus, Reg. No. 22,466; William I. Solomon, Reg. No. 28,565; Gregory E. Montone, Reg. No. 28,141; Ronald J. Shore, Reg. No. 28,577; Donald E. Stout, Reg. No. 26,422; Alan E. Schiavelli, Reg. No. 32,087; James N. Dresser, Reg. No. 22,973 and Carl I. Brundidge, Reg. No. 29,621 to prosecute and transact all business connected with this application and any related United States application and international applications. Please direct all communications to the following address:

Antonelli, Terry, Stout & Kraus
Suite 1800
1300 North Seventeenth Street
Arlington, Virginia 22209
Telephone: (703) 312-6600
Fax: (703) 312-6666

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United State Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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Date _____ Inventor _____
Residence _____ Citizenship _____
Post Office Address _____
Date _____ Inventor _____
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Residence _____ Citizenship _____
Post Office Address _____
Date _____ Inventor _____
Residence _____ Citizenship _____
Post Office Address _____

DECLARATION AND POWER OF ATTORNEY FILED WITH U.S. DESIGNATED OFFICE UNDER 35 U.S.C. 371(c)(4)

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name, I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

ELECTROLYTE REBALANCING SYSTEM

the specification of which was filed as PCT International Application No. PCT/GB00/02536
filed 30th June, 2000 and was amended on _____ (if applicable)

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

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I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)

Priority Claimed

<u>PCT/GB00/02103</u> (Number)	<u>GB</u> (Country)	<u>2/7/1999</u> (Day/Month/Year Filed)	<input checked="" type="checkbox"/> Yes	<input type="checkbox"/> No
<u>9928344.2</u> (Number)	<u>GB</u> (Country)	<u>30/11/1999</u> (Day/Month/Year Filed)	<input checked="" type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.57(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

_____ (Application Serial No.)	_____ (Filing Date)	_____ (Status: patented, pending, abandoned)
_____ (Application Serial No.)	_____ (Filing Date)	_____ (Status: patented, pending, abandoned)
_____ (Application Serial No.)	_____ (Filing Date)	_____ (Status: patented, pending, abandoned)
_____ (Application Serial No.)	_____ (Filing Date)	_____ (Status: patented, pending, abandoned)

I hereby appoint as principal attorneys: Donald R. Antonelli, Reg. No. 20.294; David T. Terry, Reg. No. 20.178; Melvin Kraus, Reg. No. 22.466; William I. Solomon, Reg. No. 28.565; Gregory E. Montone, Reg. No. 28.141; Ronald J. Shore, Reg. No. 28.577; Donald E. Stout, Reg. No. 26.422; Alan E. Schiavelli, Reg. No. 32.087; James N. Dresser, Reg. No. 22.973 and Carl I. Brundage, Reg. No. 29.621 to prosecute and transact all business connected with this application and any related United States application and international applications. Please direct all communications to the following address:

Antonelli, Terry, Stout & Kraus
Suite 1800
1300 North Seventeenth Street
Arlington, Virginia 22209
Telephone: (703) 312-6600
Fax: (703) 312-6666

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

(Full Name)

(Signature)

Date _____ Inventor Patrick John MORRISSEY
Residence United Kingdom Citizenship an Irish Citizen
Post Office Address 70 Newtown Road, New Denham, UB9 4BD, United Kingdom
Date 1 March 2002 Inventor Philip John MITCHELL
Residence United Kingdom Citizenship a British Citizen
Post Office Address 1 Crompton Close, Loughborough, LE1 13SF, United Kingdom
Date _____ Inventor Stewart Ernest MALE
Residence United Kingdom Citizenship a British Citizen
Post Office Address 68 Farm Close, East Grinstead, West Sussex, RH19 3HQ, UK
Date _____ Inventor _____
Residence _____ Citizenship _____
Post Office Address _____
Date _____ Inventor _____
Residence _____ Citizenship _____
Post Office Address _____
Date _____ Inventor _____
Residence _____ Citizenship _____
Post Office Address _____
Date _____ Inventor _____
Residence _____ Citizenship _____
Post Office Address _____
Date _____ Inventor _____
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Post Office Address _____
Date _____ Inventor _____
Residence _____ Citizenship _____
Post Office Address _____
Date _____ Inventor _____
Residence _____ Citizenship _____
Post Office Address _____



DECLARATION AND POWER OF ATTORNEY FILED WITH U.S. DESIGNATED OFFICE UNDER 35 U.S.C. 371(c)(4)

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name, I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

ELECTROLYTE REBALANCING SYSTEM

the specification of which was filed as PCT International Application No. PCT/GB00/02536
filed 30th June, 2000 and was amended on _____ (if applicable)

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)

Priority Claimed

<u>PCT/GB00/02103</u> (Number)	<u>GB</u> (Country)	<u>2/7/1999</u> (Day/Month/Year Filed)	<input checked="" type="checkbox"/> Yes	<input type="checkbox"/> No
<u>9928344.2</u> (Number)	<u>GB</u> (Country)	<u>30/11/1999</u> (Day/Month/Year Filed)	<input checked="" type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No

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_____ (Application Serial No.)	_____ (Filing Date)	_____ (Status: patented, pending, abandoned)
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_____ (Application Serial No.)	_____ (Filing Date)	_____ (Status: patented, pending, abandoned)
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I hereby appoint as principal attorneys; Donald R. Antonelli, Reg. No. 20,296; David T. Terry, Reg. No. 20,178; Melvin Kraus, Reg. No. 22,466; William I. Solomon, Reg. No. 28,565; Gregory E. Montone, Reg. No. 28,141; Ronald J. Shore, Reg. No. 28,577; Donald E. Stout, Reg. No. 26,422; Alan E. Schiavelli, Reg. No. 32,087; James N. Dresser, Reg. No. 22,973 and Carl I. Brundidge, Reg. No. 29,621 to prosecute and transact all business connected with this application and any related United States application and international applications. Please direct all communications to the following address:

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	(Full Name)	(Signature)
Date _____	Inventor Patrick John MORRISSEY	
Residence United Kingdom		Citizenship an Irish Citizen
Post Office Address 70 Newtown Road, New Denham, UB9 4BD, United Kingdom		
Date _____	Inventor Philip John MITCHELL	
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Post Office Address 1 Crompton Close, Loughborough, LE1 13SF, United Kingdom		
Date 14 March 2002	Inventor Stewart Ernest MALE	S.E. Male
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Date _____	Inventor _____	
Residence _____		Citizenship _____
Post Office Address _____		
Date _____	Inventor _____	
Residence _____		Citizenship _____
Post Office Address _____		
Date _____	Inventor _____	
Residence _____		Citizenship _____
Post Office Address _____		
Date _____	Inventor _____	
Residence _____		Citizenship _____
Post Office Address _____		
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Residence _____		Citizenship _____
Post Office Address _____		
Date _____	Inventor _____	
Residence _____		Citizenship _____
Post Office Address _____		